

# A THREE-PHASE HYDRODYNAMIC MODEL FOR SLURRY REACTORS

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## ABSTRACT

A predictive three-phase hydrodynamic model for slurry-bubble column reactors has been developed from the point of view of kinetic theory of granular flow (Gidaspow, 1994a). The model is based on first principles method and can provide a complete description of the flow field. To experimentally validate the code, local gas holdups have been measured at the National Energy Technology Laboratory (NETL) by inserting a dual conductivity probe horizontally into the reactor at various locations. Predicted gas holdups are in reasonable agreement with the experimental results in the bulk region of the reactor.

## INTRODUCTION

Slurry-Bubble-Column Reactors (SBCRs) are the preferred reactor type of synthesis gas conversion. They are flexible and may be tailored to produce high quality transportation fuel and a variety of products. Advantages of SBCRs over conventional reactors such as fixed bed include efficient heat transfer, high conversions, high olefin yields, use of low  $H_2/CO$  ratio synthesis gas, high flexibility. Despite these advantages, the Fischer-Tropsch process involving the SBCR has never been able to compete economically with conventional petroleum-base fuels. Thus, it is essential to improve its performance. Most of the studies have been concentrated on the development and optimization of new catalysts and improvement of known catalysts (Kitzelmann et al., 1977; Kolbel and Tillmetz, 1976; Zein et al., 1978). The understanding of the hydrodynamics in SBCR has received only little attention until now (Zhou, 1993) though required for a reliable design and scaleup of Fischer-Tropsch slurry reactors. Existing models such as dispersion, two fluid models, though useful starting

points for scaleup, cannot produce an adequate picture of the flow dynamics at industrially-relevant conditions of high temperatures and pressures and in the presence of real reaction fluids. Recent thorough reviews show that there exist no such models (hydrodynamic and Fisher-Tropsch reaction kinetics) in the open literature. The overall objective of our research is to develop design models for slurry-bubble-column reactors and gain an understanding of reacting fluid dynamics at high temperatures and pressures.

In view of the complexity of such multiphase reactive models, a hydrodynamic model is initially recommended. This paper presents a predictive three-phase hydrodynamic model for SBCRs. The model uses the conservation of mass, momentum, and energy for each phase. The catalyst phase pressure due to particle collision and its viscosity are derived from the kinetic theory of granular flow and are expressed in terms of the granular temperature. The latter is computed from a balance of the random kinetic energy which is similar to the well-known  $k - \epsilon$  model used in single phase flow and extended to multiphase systems by Ahmadi and Ma (1990) and Cao and Ahmadi (1995). Predicted gas holdups in the bulk region reasonably agree with the data.

## NOMENCLATURE

$C_D$	drag coefficient
$C_{Pg}$	specific heat of gas
$C_{Pp}$	specific heat of phase p
$d_p$	diameter of particle or droplet
$e_p$	restitution coefficient of phase p

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$G_g$	forces per unit mass on gas phase
$G_p$	forces per unit mass on phase p
$H_g$	enthalpy of gas phase
$H_p$	enthalpy of phase p
$h_{vp}$	gas-phase p heat transfer coefficient
$g$	gravity acceleration
$g_{0p}$	radial distribution function of phase p
$[I]$	unit tensor
$k_g$	thermal conductivity of gas
$k_p$	thermal conductivity of phase p
$k_g^0$	mean thermal conductivity of gas
$k_p^0$	mean thermal conductivity of phase p
$k_p^*$	effective thermal conductivity of phase p
$Pr$	Prandtl number
$R$	universal gas constant
$Re_p$	Reynolds number based on $d_p$
$T$	thermal temperature
$T_g$	gas thermal temperature
$T_p^0$	thermal temperature of phase p
$T_g^0$	reference gas temperature
$T_p^0$	reference temperature of phase p
$u$	velocity in the x direction
$v$	velocity in the y direction
$z$	compressibility factor

Greek letters:

$b_{pl}$	frictional coefficient between phase p and l
$g_p$	collisional energy dissipation
$e_k$	volume fraction of phase k
$\Theta$	granular temperature
$k_p$	conductivity of fluctuating energy
$m_p$	viscosity of phase p
$x_p$	bulk viscosity
$r_p$	density of phase p
$t_g$	gas-phase shear stress
$t_p$	shear stress of phase p
$\Psi_p$	sphericity of particle or droplet

*Subscripts*

g, l, s	gas, liquid, solid respectively
k	gas, liquid, solid (g,l,s)
p	solid or liquid phase (l,s)

*Superscript*

T	transpose
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Hydrodynamics model for three-phase flow

For transient, three-phase flow the general mass conservation equations and separate phase momentum equations are written as follows in conservation law form for hydrodynamic model B (Bouillard et al. [1989], Gidaspow, [1994b], and Gamwo *et al.*, [1995, 1999]):

Conservation equations :

Continuity equation for gas, liquid and solid [k=g,l,s]:

$$\frac{\partial}{\partial t}(\mathbf{e}_k \mathbf{r}_k) + \nabla \cdot (\mathbf{e}_k \mathbf{r}_k \bar{\mathbf{v}}_k) = 0 \quad (1)$$

Conservation of phase-volume fractions:

$$\mathbf{e}_g + \mathbf{e}_l + \mathbf{e}_s = 1 \quad (2)$$

Gas-phase momentum equation :

$$\begin{aligned} \frac{\partial}{\partial t}(\mathbf{e}_g \mathbf{r}_g \bar{\mathbf{v}}_g) + \nabla \cdot (\mathbf{e}_g \mathbf{r}_g \bar{\mathbf{v}}_g \bar{\mathbf{v}}_g) &= \mathbf{e}_g \mathbf{r}_g \bar{\mathbf{G}}_g + \nabla \cdot [\bar{\mathbf{t}}_g] \\ &+ \sum_{p=l,s} \mathbf{b}_{gp}(\bar{\mathbf{v}}_p - \bar{\mathbf{v}}_g) \quad (3) \end{aligned}$$

where  $\mathbf{b}_{gp}$  are the drag coefficients between the gas and the dispersed phases (l,s).  $\bar{\mathbf{t}}_g$  is the gas-phase stress tensor.

Liquid-phase momentum equation :

$$\begin{aligned} \frac{\partial}{\partial t}(\mathbf{e}_l \mathbf{r}_l \bar{\mathbf{v}}_l) + \nabla \cdot (\mathbf{e}_l \mathbf{r}_l \bar{\mathbf{v}}_l \bar{\mathbf{v}}_l) &= \mathbf{e}_l \mathbf{r}_l \bar{\mathbf{G}}_l + \nabla \cdot \bar{\mathbf{t}}_l \\ &+ \mathbf{b}_{lg}(\bar{\mathbf{v}}_g - \bar{\mathbf{v}}_l) + \mathbf{b}_{ls}(\bar{\mathbf{v}}_s - \bar{\mathbf{v}}_l) \quad (4) \end{aligned}$$

Solid- phase momentum equation :

$$\begin{aligned} \frac{\partial}{\partial t}(\mathbf{e}_s \mathbf{r}_s \bar{\mathbf{v}}_s) + \nabla \cdot (\mathbf{e}_s \mathbf{r}_s \bar{\mathbf{v}}_s \bar{\mathbf{v}}_s) &= \mathbf{e}_s \mathbf{r}_s \bar{\mathbf{G}}_s \\ &+ \nabla \cdot \bar{\mathbf{t}}_s + \mathbf{b}_{sg}(\bar{\mathbf{v}}_g - \bar{\mathbf{v}}_s) + \mathbf{b}_{ls}(\bar{\mathbf{v}}_l - \bar{\mathbf{v}}_s) \quad (5) \end{aligned}$$

Gas-phase energy equation in terms of enthalpy H is:

$$\begin{aligned} \frac{\partial}{\partial t}(\mathbf{e}_g \mathbf{r}_g H_g) + \nabla \cdot (\mathbf{e}_g \mathbf{r}_g H_g \bar{\mathbf{v}}_g) &= -\nabla \cdot (q_g) \\ &+ \left( \frac{\partial P_g}{\partial t} + P_g \nabla \cdot \bar{\mathbf{v}}_g \right) \\ &+ \sum_{p=l,s} \left[ \mathbf{b}_{gp}(\bar{\mathbf{v}}_p - \bar{\mathbf{v}}_g)^2 + h_{vp}(T_p - T_g) \right] \quad (6) \end{aligned}$$

where  $h_{vp}$  is the heat transfer coefficient and  $q_g$  is the gas-phase conductive heat transfer described by Fourier's law  $q_g = k_g \nabla T_g$

Dispersed-phases energy equations (p=l,s):

$$\begin{aligned} \frac{\partial}{\partial t}(\mathbf{e}_p \mathbf{r}_p H_p) + \nabla \cdot (\mathbf{e}_p \mathbf{r}_p H_p \dot{\mathbf{v}}_p) = \\ -\nabla \cdot (q_p) + h_{vp}(T_g - T_p) \\ + \sum_{p=l,s} \mathbf{b}_{pg} \left( \vec{\mathbf{v}}_p - \vec{\mathbf{v}}_g \right)^2 \quad (7) \end{aligned}$$

Fluctuation energy equation for the particles:

Following the approach of Ahmadi and Ma (1990) and Cao and Ahmadi (1995), we assume that the fluctuating velocity of particles is the same as that of the fluid. Hence only the equation for the granular temperature  $\Theta_s$  of the particles is needed.

$$\begin{aligned} \frac{3}{2} \left[ \frac{\partial}{\partial t} (\mathbf{e}_s \mathbf{r}_s \Theta_s) + \nabla \cdot (\mathbf{e}_s \mathbf{r}_s \Theta_s \mathbf{n}_s) \right] = \nabla \cdot (\mathbf{k}_s \nabla \Theta_s) \\ - \mathbf{g}_s + \left[ \mathbf{t}_s \right] : \nabla \vec{\mathbf{v}}_s \quad (8) \end{aligned}$$

Where  $\mathbf{k}_s$  is the dense-phase diffuse coefficient for granular temperature, and  $\mathbf{g}_s$  is the dissipation due to collision.

Constitutive Equations:

The most important part of the kinetic theory needed in the hydrodynamic model is the computation of the particulate viscosity. The expression we use for the viscosity was verified to give the viscosity measured by classical methods for gas-solid (Gidaspow and Huilin, 1996; Buyevich and Cody, 1998) and for liquid-solid (Gidaspow and Huilin, 1998b) fluidization. The major difference in kinetic theory between fluidization of particles in air and in water is that the particles do not contact each other during the collision. In liquids there exists a film between the particles that gives rise to a lubrication force. Thus in applying the kinetic theory to liquids one must take the restitution coefficient to be unity and lump all the dissipation into an effective drag in the fluctuating energy equation.

Interphase drag coefficients: (p=l,s)

-Based on Ergun equation for  $\mathbf{e}_g < 0.8$

$$\mathbf{b}_{gp} = \mathbf{b}_{pg} = 150 \frac{(1 - \mathbf{e}_g) \mathbf{e}_p \mathbf{m}_g}{(\mathbf{e}_g d_p \mathbf{y}_p)^2} + 1.75 \frac{\mathbf{r}_g \mathbf{e}_p / \vec{\mathbf{v}}_g - \vec{\mathbf{v}}_p /}{\mathbf{e}_g d_p \mathbf{y}_p} \quad (9)$$

-Based on Wen and Yu's expression for  $\mathbf{e}_g \geq 0.8$

$$\mathbf{b}_{gp} = \mathbf{b}_{pg} = \frac{3}{4} C_D \frac{\mathbf{r}_g \mathbf{e}_p / \vec{\mathbf{v}}_g - \vec{\mathbf{v}}_p /}{d_p \mathbf{y}_p} \mathbf{e}_g^{-2.65} \mathbf{e}_g < 0.8 \quad (10)$$

where the drag coefficient  $C_D$  is given by

$$C_D = \frac{24}{Re_p} [1 + 0.15 Re_p^{0.687}] \quad \text{if } Re_p > 1000$$

$$C_D = 0.44 \quad \text{if } Re_p \leq 1000$$

$$\text{and } Re_p = \frac{\mathbf{e}_g \mathbf{r}_g / \vec{\mathbf{v}}_g - \vec{\mathbf{v}}_p /}{d_p \mathbf{y}_p} \frac{\mathbf{m}_g}{\mathbf{m}_g}$$

-Liquid-solid drag coefficient

$$\mathbf{b}_{ls} = \mathbf{b}_{sl} = \frac{3}{2} (1 + e) \frac{\mathbf{r}_s \mathbf{r}_l \mathbf{e}_s \mathbf{e}_l (d_s + d_g)^2}{\mathbf{r}_s d_s^3 + \mathbf{r}_l d_l^3} \left| \vec{\mathbf{v}}_l - \vec{\mathbf{v}}_s \right| \quad (11)$$

Shear stresses:

$$[\mathbf{t}_g] = \left\{ -P_g - \frac{2}{3} \mathbf{m}_g \mathbf{e}_g \nabla \cdot \vec{\mathbf{v}}_g \right\} [I] + \mathbf{m}_g \mathbf{e}_g \left[ \nabla \vec{\mathbf{v}}_g + (\nabla \vec{\mathbf{v}}_g)^T \right] \quad (13)$$

$$[\mathbf{t}_p] = \left\{ -P_p + \left( \mathbf{x}_p - \frac{2}{3} \mathbf{m}_p \right) \nabla \cdot \vec{\mathbf{v}}_p \right\} [I] + \mathbf{m}_p \left[ \nabla \vec{\mathbf{v}}_p + (\nabla \vec{\mathbf{v}}_p)^T \right] \quad (14)$$

where

Particulate viscosity:

$$\mathbf{m}_p = \frac{2 \mathbf{m}_{p,dil}}{(1 + e_p) g_{0p}} \left\{ 1 + \frac{4}{5} (1 + e_p) g_{0p} \mathbf{e}_s \right\}^2 + \frac{4}{5} \mathbf{e}_p^2 \mathbf{r}_p d_p g_{0p} (1 + e_p) \sqrt{\frac{\Theta_p}{\mathbf{p}}}$$

$$\mathbf{m}_{p,dil} = \frac{5}{96} \mathbf{r}_p d_p \sqrt{\mathbf{p} \Theta_p} \quad \text{dilute phase viscosity}$$

Bulk viscosity:

$$\mathbf{x}_p = \frac{4}{3} \mathbf{e}_p^2 \mathbf{r}_p d_p g_{0p} (1 + e_p) \sqrt{\frac{\Theta_p}{\mathbf{p}}}$$

$$g_{0p} = \left\{ 1 - \left( \frac{\mathbf{e}_p}{\mathbf{e}_{p,max}} \right)^{1/3} \right\}^{-1} \quad \text{radial distribution function}$$

Equation of state for particles (p=s)

$$P_p = \mathbf{r}_p \mathbf{e}_p \Theta_p \{1 + 2(1 + e_p) g_{0p} \mathbf{e}_p\} \quad (15)$$

Equation of state for gas:

$$\mathbf{r}_g = \frac{\bar{M}_g P_g}{zRT_g} \text{ where } \bar{M}_g \text{ is the average gas molecular weight}$$

and z is the compressibility factor

Collisional energy dissipation:

$$\mathbf{g}_p = 3(1 - e_p^2) \mathbf{e}_p^2 \mathbf{r}_p g_{0p} \Theta_p \left\{ \frac{4\sqrt{\Theta_p}}{d_p \sqrt{\mathbf{p}}} - \nabla \cdot \vec{v}_p \right\} \quad (16)$$

Dense-phase conductivity:

$$\mathbf{k}_p = \frac{2k_{p,dil}}{(1 + e_p) g_{0p}} \left\{ 1 + \frac{6}{5} (1 + e_p) g_{0p} \mathbf{e}_p \right\}^2 + 2\mathbf{e}_p^2 \mathbf{r}_p d_p g_{0p} (1 + e_p) \sqrt{\frac{\Theta_p}{\mathbf{p}}} \quad (17)$$

Where the dilute phase conductivity is

$$\mathbf{k}_{p,dil} = \frac{75}{384} \mathbf{r}_p d_p \sqrt{\mathbf{p} \Theta_p}$$

Gas thermal conductivity:

$$k_g = (1 - \sqrt{1 - \mathbf{e}_g}) k_g^0$$

Dispersed-phase heat transfer:

$$k_p = \frac{\sqrt{\mathbf{e}_s}}{(1 - \mathbf{e}_g)} \left\{ \mathbf{j} \frac{k_p^*}{k_g^0} + (1 - \mathbf{j}) \frac{k_p^0}{k_g^0} \right\} k_g^0 \quad (18)$$

where

$$\frac{k_p^0}{k_g^0} = \frac{2}{A_p} \left\{ \frac{B_p \left\{ (k_p^* / k_g^0) - 1 \right\}}{A_p^2 (k_p^* / k_g^0)} \cdot \ln \left( \frac{k_p^* / k_g^0}{B_p} \right) - \frac{B_p - 1}{A_p} - \frac{B_p + 1}{2} \right\}$$

and

$$A_p = 1 - \frac{B_p}{\left( \frac{k_p^*}{k_g^0} \right)}$$

$$B_p = 1.25 \left( \frac{\mathbf{e}_p}{\mathbf{e}_g} \right)^{10/9} (1 + 3\mathbf{c})$$

$$\mathbf{c} = \sqrt{\frac{(\sum \mathbf{e}_p \mathbf{r}_p)(\sum \mathbf{e}_p \mathbf{r}_p / d_p^2)}{(\sum \mathbf{e}_p \mathbf{r}_p / d_p)^2} - 1}$$

$$k_p^* = 0.33 \quad \mathbf{j} = 7.3 \times 10^{-3}$$

$$p = l, s$$

External forces acting on each phase:

$$G_g = \frac{g}{\mathbf{e}_g} \quad (19)$$

$$G_p = \frac{g}{\mathbf{e}_g} \left( 1 - \frac{1}{\mathbf{r}_p} \sum_{k=g,l,s} \mathbf{e}_k \mathbf{r}_k \right) \quad p = l, s \quad (20)$$

Enthalpy:

$$H_g = C_{Pg} (T_g - T_g^0) \quad (21)$$

$$H_p = C_{Pp} (T_p - T_p^0) \quad (22)$$

Gas-solid heat transfer:

$$h_{vp} = Nu_p \frac{k_g^0}{d_p} \quad (23)$$

where

$$Nu_p = \left\{ \left( 2 + 5\mathbf{e}_p^2 \right) \left( 1 + 0.7 \text{Re}_p^{0.2} \text{Pr}^{1/3} \right) + \left( \frac{2}{15} + 1.2\mathbf{e}_p^2 \right) \text{Re}_p^{0.7} \text{Pr}^{1/3} \right\} Sp_p$$

$$\text{and } \text{Pr} = \frac{C_{Pg} \mathbf{m}_g}{k_g^0} ; \quad Sp_p = \frac{6\mathbf{e}_p}{d_p} ; \quad k_g^0 = 8.65 \times 10^5 \left( \frac{T_g}{1400} \right)^{1.786}$$

Numerical Scheme

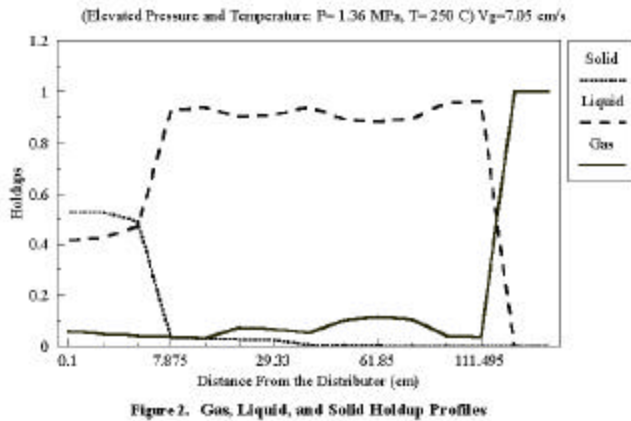
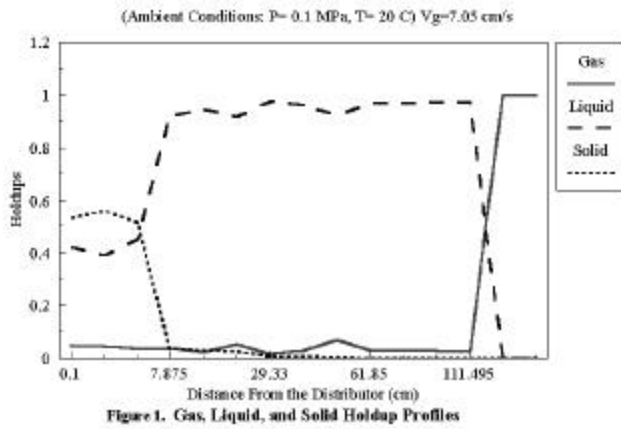
The governing equations along with the constitutive equations are solved for  $\mathbf{e}_g, \mathbf{e}_l, \mathbf{e}_s, \mathbf{v}_g, \mathbf{v}_l, \mathbf{v}_s, T_g, T_l, T_s$ , and  $\mathbf{P}_g$  using the Implicit Continuous Eulerian (ICE) method (Rivard and Torrey, 1977; Jayaswal, 1991, Gamwo et al., 1999) with appropriate initial and boundary conditions. The computations are carried out using a mesh of finite-difference cells fixed in two-dimensional space (Eulerian mesh). The scalar variables are located at the cell center and the vector variables at the cell boundaries. The momentum equation is solved using staggered mesh, where as for continuity equation a donor cell method was used, (Gamwo, et al., 1999).

The partial differential equations are well posed (Gamwo, et al., 1995). The time step is chosen to satisfy the Courant stability criterion (Courant, et al., 1952). The numerical stability of the equations can be obtained using the Von Neumann stability analysis, as illustrated by Lyczkowski, et al. (1978) and Prosperetti (1982).

## Computational results

Nonuniform finite difference grids were used. Axial symmetry was assumed. The reactor was finite differenced into 14x94 computational cells. Cells sizes in the axial direction vary from 0.2 to 12.70 cm and from 0.1 to 0.4 cm in the radial direction. Numerical computations were carried out for 1.8 sec. with a time increment of  $2 \times 10^{-6}$  sec.

To show the wide range of the model applicability, profiles of gas/liquid/solid holdups were computed at both laboratory (ambient temperature and pressure) and industrially-relevant (high temperature and pressure) conditions. Figures 1 and 2 show the three phases holdup profiles at different reactor heights at ambient and industrially-relevant conditions respectively.



Both figures clearly demonstrate that the solids concentration at the bottom of the reactor is very high and decreases sharply with reactor height. The solids holdup at the top of the reactor is nearly zero implying the absence of catalyst in that region. The liquid holdup at the top of the reactor is high, due to the

relative lack of solids. Overall the solids holdup decreases with height, the liquid holdup increases with height and the gas holdup remains fairly constant throughout the reactor

## Experimental gas holdups

The acquisition and analysis of experimental data are not repeated here as they are summarized by Soong et al., (1997). Experiments were conducted in a 10.16 cm I.D. x 243.84 cm reactor operating in the batch-mode with stationary Drakeol-10 oil/90  $\mu$ m glass beads and a continuous flow of nitrogen gas. The column has 12 different axial locations for data collection. The local gas holdups were measured at NETL by inserting a dual conductivity probe (hot wire) horizontally into the column at any of the 12 positions and moving it to the desired radial position. The holdups were measured at three locations above the gas distributor in the bubble column. The experiments were conducted at both laboratory and industrially-relevant conditions at superficial gas velocity of 7.05 cm/s with Drakeol-10 oil as the liquid phase, nitrogen as the gas phase and 90  $\mu$ m glass beads as the solid phase medium.

## Comparison of model results with experiments

Holdups measurement is a sensitive test of the theory, since it is not prescribed at any of the boundaries in the simulation of the slurry-bubble column reactor. Hence, measured gas holdups were compared with simulated results under conditions shown in Table 1 that closely resemble the experimental conditions.

Table 1. Simulation Conditions

Fluids	Drakeol-10 oil/Nitrogen
Temperatures, $^{\circ}$ C	20 and 250
Pressures, MPa	0.1 and 1.36
Superficial gas velocity, cm/s	7.05
Particles	Glass beads
Particle mean diameter, cm	0.009
Particle density, g/cm <sup>3</sup>	2.362
Particle sphericity	1.0

Figure 3 shows a comparison of the experimentally determined gas holdup for slurry of 90  $\mu$ m glass beads/Drakeol- 10/nitrogen to computations using our three-phase model.

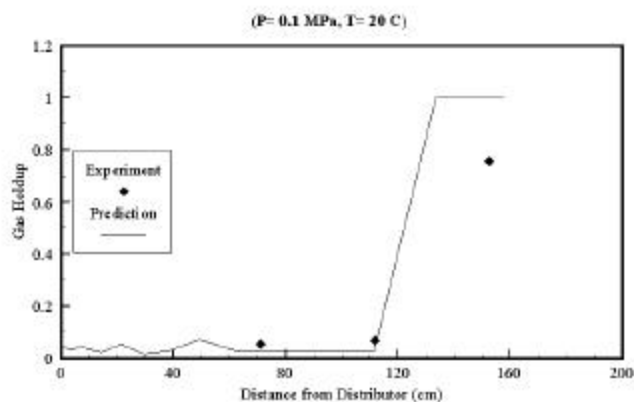


Figure 3. Predicted and Experimental Gas Holdup Vs Reactor Height

Both the experiment and the theory show low and nearly-constant gas holdup in the bottom and middle regions of the reactor, then the gas holdup rises sharply in the free-board region. The agreement between the experiment and the model is reasonable, except in the disengagement zone. At this location, the experiment showed a lower gas holdup probably caused by asymmetric motion of the bubble. This larger difference may be due to the symmetry assumption and to the absence of the third dimension.

## Conclusions

A three-phase reactor model for describing the hydrodynamics in slurry-bubble column has been developed. Computed gas holdups at atmospheric pressure and temperature agree well with experimental data except in the disengagement region. This discrepancy is likely due to the symmetry assumption which is not observed experimentally and to the third dimension. To correct this deficiency we are in the process of developing a three-dimensional code with no symmetry assumption.

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